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WO 1998/028070 A1

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(58) Field of Search

UK CL (Edition V) **B1W**

INT CL⁷ **B01D, B01J, F01N, F02D**

Other: **ONLINE : EPODOC, JAPIO, WPI.**

(54) Abstract Title

A selective catalytic reduction device and methods for use therein

(57) A selective catalytic reduction system for engine exhaust injects a solution comprising a source of ammonia, preferably an aqueous urea solution, or other ammonia solution, to preferably evaporate, decompose and hydrolyze to produce ammonia to react with and reduce NO_x in the exhaust. An inertial impactor (32) in the housing between the injector (22) and the catalyst (28) is impacted by the solution droplets and holds same until evaporated, decomposed and hydrolyzed to ammonia.

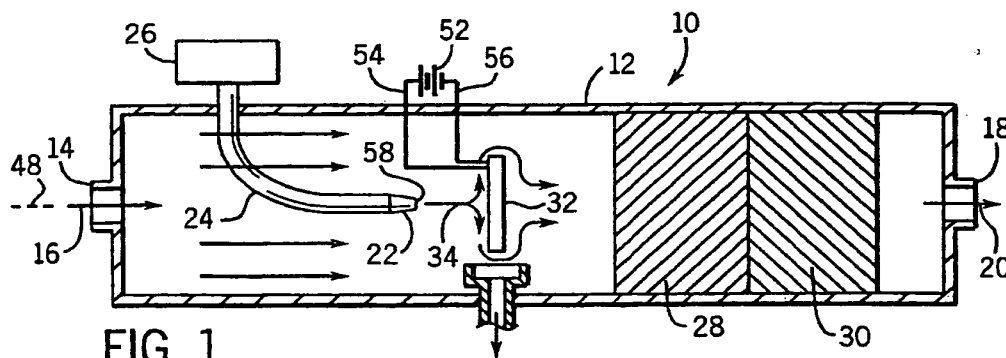


FIG. 1

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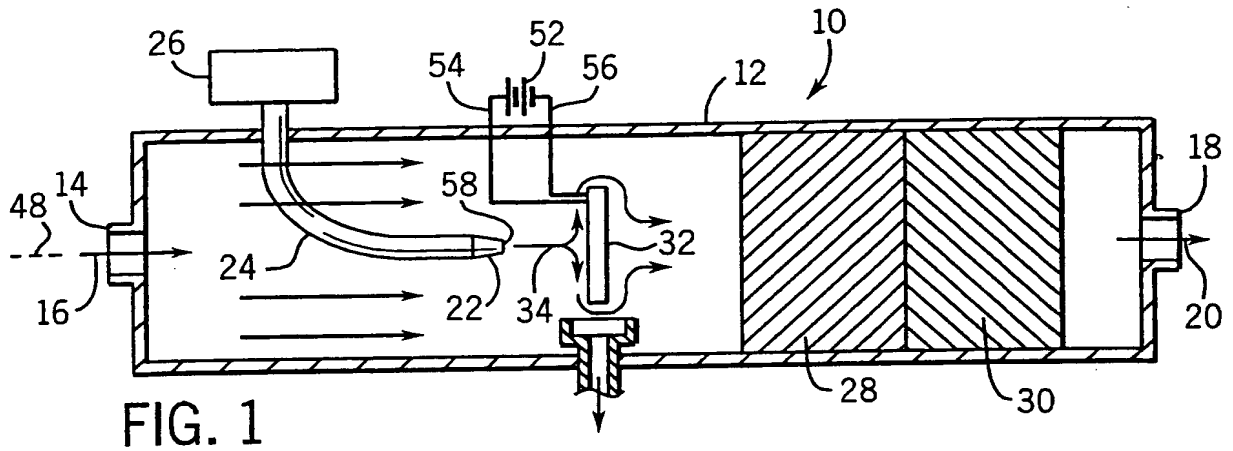


FIG. 1

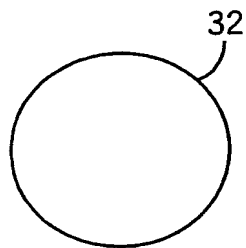


FIG. 2

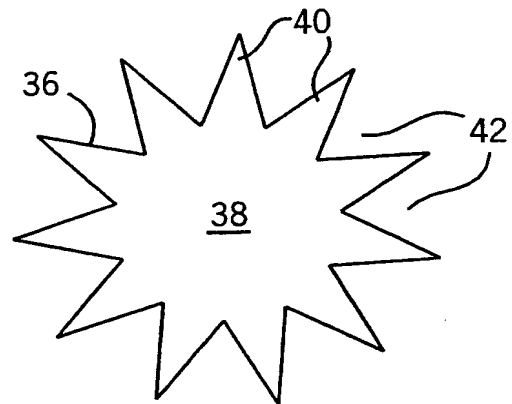


FIG. 3

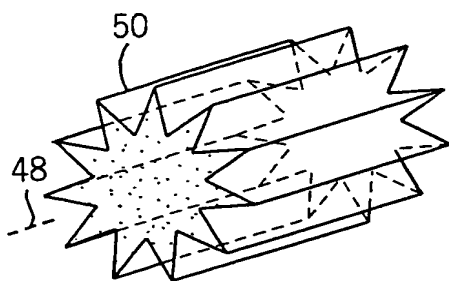


FIG. 4

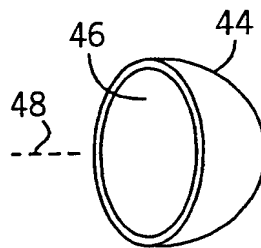


FIG. 5

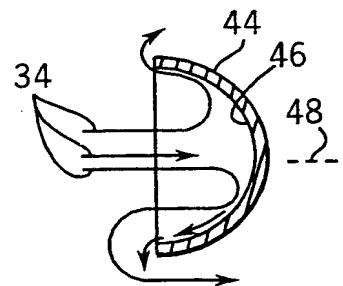


FIG. 6

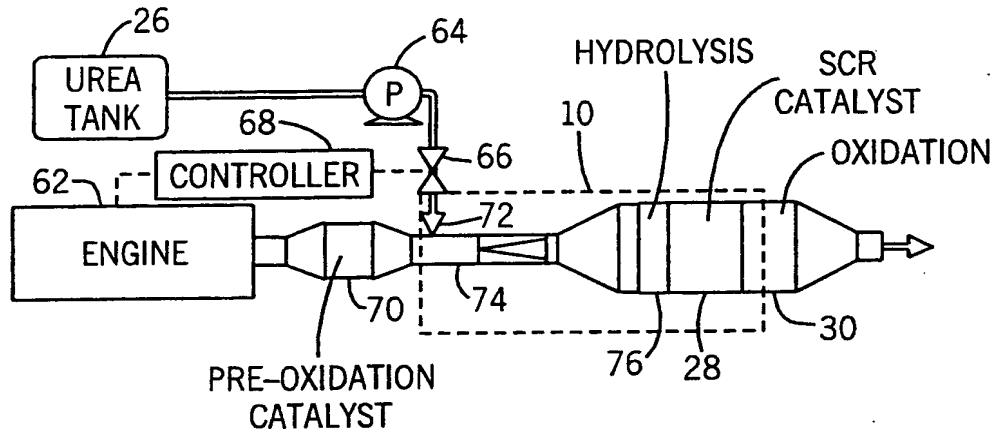


FIG. 7

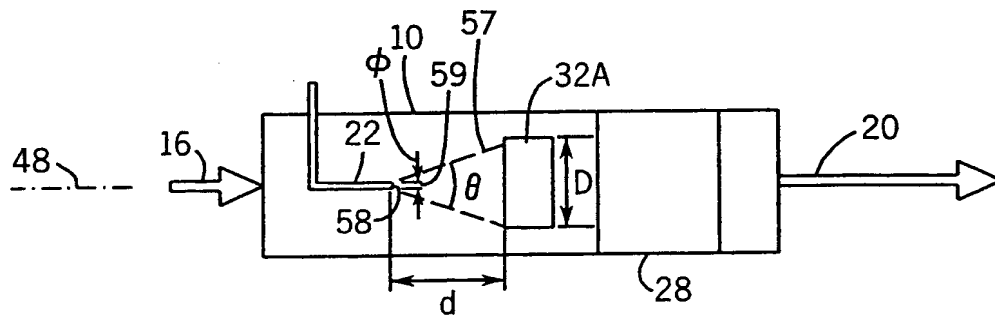


FIG. 8

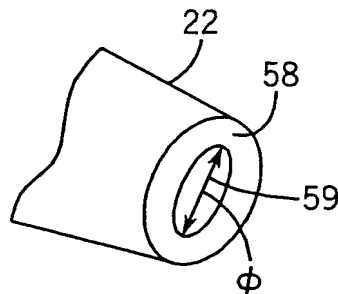


FIG. 9

BACKGROUND AND SUMMARY

5 The invention relates to selective catalytic reduction systems for internal combustion engine exhaust, including for use in industrial processes and in mobile and stationary diesel, natural gas, and other engine applications.

10 Selective catalytic reduction (SCR) of nitrogen oxides (NO_x) with urea or ammonia solution is known in industrial processes as well as in stationary diesel engine applications. In the SCR system, urea or ammonia solution is injected into the exhaust gas flow stream before the catalyst. Water in the droplets from the injection solution evaporates, leaving urea behind, which decomposes and hydrolyzes into ammonia, i.e. ammonia is produced through the process of droplet evaporation and urea chemical decomposition and hydrolysis. NO_x reacts with the ammonia and is catalytically reduced in the presence of such ammonia.

15 Thus far, SCR catalysts have not been commercially used for mobile applications because of complexity. One of the many obstacles is that the limited space available in mobile applications makes it hard to evaporate droplets and decompose and hydrolyze the urea completely and achieve a high enough ammonia to NO_x ratio. At low diesel engine exhaust temperatures and high gas flow velocities (e.g. about 20 meters per second), a distance of several meters (i.e. a time of 0.1 to 0.3 seconds) between the injector and the catalyst is required for the aqueous urea solution spray to vaporize, for urea to decompose and hydrolyze into ammonia and carbon dioxide, and for the ammonia to become uniformly distributed across the flow front before entering the catalyst. In order to be practical, this distance must be shortened to less than about 0.5 meters. The droplet evaporation and urea decomposition and hydrolysis are key factors affecting performance of the SCR system.

25 The present invention provides a selective catalytic reduction system which may be used in mobile applications. The noted distance between the injector and the catalyst is shortened by retaining the solution on an impactor while evaporation, and decomposition in the case of urea, occur. The solution evaporation rate, and decomposition rate in the case of urea, is further enhanced through heating. The invention also includes an embodiment that enhances the hydrolysis process.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a side schematic sectional view of a selective catalytic reduction, SCR, device in accordance with an embodiment of the invention.

5 Fig. 2 is an end elevation view of a component of Fig. 1.

Fig. 3 is like Fig. 2 and shows another embodiment.

Fig. 4 is a perspective view of an alternate component of Fig. 1.

Fig. 5 is a perspective view of another alternate component of Fig. 1.

Fig. 6 is a side sectional view of the component of Fig. 5.

10 Fig. 7 is a schematic illustration of an SCR, selective catalytic reduction, system.

Fig. 8 is similar to Fig. 1 and further illustrates an embodiment in accordance with the invention.

Fig. 9 is an isometric enlarged view of a portion of Fig. 8.

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DETAILED DESCRIPTION

Fig. 1 shows a selective catalytic reduction device 10 for internal combustion engine exhaust. The device includes a housing 12 having an upstream inlet 14 for receiving exhaust as shown at arrow 16, and a downstream outlet 18 for discharging the exhaust as shown at arrow 20. An injector 22 in the housing injects aqueous urea solution through tubular conduit 24 from reservoir or tank 26. As known, the droplets in the solution evaporate, and the urea decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in the exhaust by catalytic reduction in the presence of such ammonia. A catalyst is provided in the housing downstream of injector 22, including SCR catalyst 28 and oxidation catalyst 30.

25 In one embodiment, a nonporous inertial impactor 32 is provided in the housing between injector 22 and catalyst 28 and is impacted by exhaust and urea flow as shown at 34. In preferred form, nonporous impactor 32 holds the solution until the droplets have evaporated and the urea has decomposed and hydrolyzed to ammonia. This is accomplished by selecting the impactor and the injection rate from the injector to satisfy the condition that the evaporation and decomposition rate at the impactor is greater than the injection rate at the injector. This is accomplished in the present invention in a number of

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ways, to be described.

In one embodiment, the noted condition is satisfied by sizing impactor 32 to a large enough size, for example a circular impactor, Fig. 2, having a large enough diameter. In embodiments where further increase in size may be objectionable because of blockage or restriction of exhaust flow, the impactor has a star shape, as shown at impactor 36, Fig. 3, with a central core 38 and a plurality of radially outwardly projecting tips 40 each providing impactation surface area yet permitting exhaust flow therebetween at spaces 42. In another embodiment, where increased lateral size is objectionable, the impactor has a bowl shape, as shown at 44, Figs. 5, 6, with a concave surface 46 facing injector 22. Concave surface 46 provides increased impactation surface area within a reduced lateral area blocking exhaust flow.

In a further embodiment, the impactor is a porous member holding the injected solution until it evaporates and decomposes. The axial length of the impactor along axis 48 may be varied to control the holding capacity of the porous impactor, for example as shown at impactor 50 in Fig. 4. In one desirable embodiment, the impactor has a rough porous collection surface, for example as disclosed in U.S. Patent 6,290,738, incorporated herein by reference, causing liquid particle separation of smaller size liquid particles than a smooth nonporous impactor impingement surface, to improve evaporation and decomposition.

A porous impactor has several advantages over a nonporous impactor. In the case of a porous impactor, it is not necessary that the urea solution evaporation and decomposition rate at the impactor be greater than the urea solution injection rate at the injector, because the impactor can be sized such that it can hold excess urea solution when the injection rate exceeds the evaporation/decomposition rate, buffering against such imbalances. In the case of a porous impactor, it may be desirable to make the impactor surface preferentially wetted by the urea solution, so that the solution will be drawn into the impactor and spread over a greater surface area, facilitating evaporation, decomposition and hydrolysis. It may be desirable to provide a high thermal conductivity impactor of low heat capacity and/or low thermal mass, because it is easier to heat, ideally by the exhaust, but also optionally by a heat source, for example as provided by a voltage source 52 external of the housing, and a pair of electrical conductors 54, 56 connecting the voltage source to the impactor. The impactor can be coated with or actually be a hydrolysis catalyst, to facilitate

decomposition.

The urea solution droplets sprayed out of urea injector 22 are impacted onto impactor 32. The impactor is placed such that the momentum of the droplets causes them to deviate from the flow stream and impact on the impactor surface. Deposited droplets are held on or within the impactor until they evaporate. For a nonporous impactor or for a porous impactor that is undersized for the injection-evaporation/decomposition imbalance, the urea solution injection rate is less than or equal to the evaporation and decomposition rate, otherwise the impactor will release larger, coalesced drops or else solid urea particles may accumulate on the impactor. The impactor is desirably heated in order to accelerate vaporization and decomposition. Depending on the urea solution injection rate and exhaust gas temperatures, heating may be done electrically or by the exhaust. The impactor should desirably have a high thermal conductivity to facilitate transfer of heat to the urea solution. If a porous impactor is used, it should be preferentially wetted by the urea solution, in order for the urea to be drawn into and held by the impactor. An advantage of heating the impactor instead of the injector is that heat transfer to the urea solution is more effective. Temporary accumulation of dry urea on the impactor, until it heats up upon engine restart, does not adversely affect performance. The axial distance along axis 48 between downstream injection tip 58 of injector 22 and catalyst 28 is less than about 0.5 meters.

In the preferred embodiment, an aqueous urea solution is injected, as described, which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in the exhaust. In further embodiments, other ammonia solutions or sources of ammonia may be used.

Fig. 7 shows an SCR system for exhaust from engine 62. Pump 64 draws aqueous urea solution from tank 26 and supplies same through valve 66 which is controlled by controller 68 which in turn is controlled by the engine, all as is known. Engine exhaust is initially supplied through pre-oxidation catalyst 70 and then mixed with the aqueous urea solution injected at 72 into exhaust pipe 74. The mixture then flows downstream to hydrolysis catalyst 76, SCR catalyst 28 and oxidation catalyst 30. In accordance with one embodiment of the present invention, the hydrolysis catalyst is removed from its location as shown at 76 and instead provided at impactor 32, Fig. 1, as shown at hydrolysis catalyst impactor 32a in Fig. 8. The impactor including the hydrolysis catalyst is spaced downstream from injector 22 and spaced upstream from SCR catalyst 28, facilitating

decomposition and hydrolysis of the aqueous urea solution at the impactor and upstream of the SCR catalyst. Housing 10 has a given cross-sectional area at SCR catalyst 28 transverse to axis 48. Hydrolysis catalyst 32a has a lateral cross-sectional area transverse to axis 48 which is less than the noted given cross-sectional area. Aqueous urea solution is injected along an injection spray angle θ at 57 from injector 22. Hydrolysis catalyst 32a has a lateral cross-sectional dimension D transverse to axis 48. Injector 22 has the noted downstream injection tip 58 spaced from hydrolysis catalyst 32a by a distance d along axis 48. Injection tip 58 has an inner diameter ϕ at 59 transverse to axis 48. In the preferred embodiment $D > 2d \tan(\theta/2) + \phi$. Hydrolysis catalyst 32a is spaced in the spray path of the aqueous urea solution, with D being larger than the diameter of the spray pattern. The described arrangement provides significant benefits over an SCR system with the conventional hydrolysis catalyst placement shown in Fig. 7. These benefits include: (1) the distance between injector 22 and SCR catalyst 28 can be shortened; (2) the size, particularly the lateral cross-sectional dimension, of the hydrolysis catalyst can be significantly reduced; (3) pressure drop is reduced because the hydrolysis catalyst no longer occupies the entire cross-sectional area of the housing as in Fig. 7; and (4) there is increased time for ammonia mixing prior to SCR catalyst 28. Hydrolysis catalyst 32a preferably has a dominant impactor effect to catch droplets of the aqueous urea solution, but in other embodiments can be a flow-through catalyst wherein some droplets would be caught by impaction, but most would be captured on the walls as they flow through the channels. The hydrolysis catalyst is preferably selected from the group consisting of: extruded material; wrapped material, such as pleated or corrugated; and layered material. The catalyst is further preferably selected from the group consisting of extruded monolith material; composite ceramic material, for example as in commonly owned co-pending U.S. patent applications Serial No. 09/573,747, filed May 18, 2000, and Serial No. 09/935,847, filed August 23, 2001; fibrous material; and metallic material, such as flow-through metal foils and the like. As in previous embodiments, the hydrolysis catalyst can be heated, e.g. by heat source 52, in addition to heating by the exhaust, to accelerate evaporation and decomposition and enhance hydrolysis.

It is recognized that various equivalents, alternatives and modifications are possible within the scope of the appended claims which distinctly claim the subject matter regarded as the invention.

CLAIMS

1. A selective catalytic reduction device for engine exhaust comprising:
a housing having an upstream inlet for receiving said exhaust, and a downstream outlet for discharging said exhaust;
an injector in said housing for injecting a solution comprising a source of ammonia to react with and reduce NO_x in said exhaust;
a catalyst in said housing downstream of said injector;
an inertial impactor in said housing between said injector and said catalyst and impacted by said exhaust and said solution.
2. A selective catalytic reduction device according to claim 1 wherein said impactor holds said solution to enhance ammonia reaction with NO_x.
3. A selective catalytic reduction device according to claim 2 wherein said source of ammonia is an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust.
4. A selective catalytic reduction device according to claim 3 wherein said impactor holds said aqueous urea solution until evaporated and decomposed to ammonia.
5. A selective catalytic reduction device according to claim 2 wherein said source of ammonia is an ammonia solution.
6. A selective catalytic reduction device according to claim 5 wherein said impactor holds said ammonia solution until evaporated.
7. A selective catalytic reduction device according to claim 2 wherein said impactor is nonporous, and wherein said impactor and the injection rate from said injector are selected to satisfy the following condition: the solution evaporation rate at said impactor is greater than the injection rate at said injector.

8. A selective catalytic reduction device according to claim 7 wherein said condition is satisfied by sizing said impactor to a large enough size.

9. A selective catalytic reduction device according to claim 7 wherein said source of ammonia is an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust, and wherein said impactor and the injection rate from said injector are selected to satisfy the following
5 condition: the aqueous urea solution evaporation and decomposition rate at said impactor is greater than the injection rate at said injector, such that said impactor holds said aqueous urea solution until evaporated and decomposed to ammonia.

10. A selective catalytic reduction device according to claim 2 wherein said impactor is porous.

11. A selective catalytic reduction device according to claim 10 wherein said impactor has a rough porous collection surface causing liquid particle separation of smaller size liquid particles than a smooth nonporous impactor impingement surface to improve evaporation of said solution.

12. A selective catalytic reduction device according to claim 11 wherein said impactor surface is preferentially wetted by said solution such that said solution is drawn into said impactor and spread over a greater surface area, enhancing evaporation.

13. A selective catalytic reduction device according to claim 10 wherein said source of ammonia is an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react and reduce NO_x in said exhaust, and wherein said impactor comprises a hydrolysis catalyst, facilitating said hydrolysis.

14. A selective catalytic reduction device according to claim 10 wherein said source of ammonia is an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust, said impactor

surface is preferentially wetted by said aqueous urea solution such that said aqueous urea solution is drawn into said impactor and spread over a greater surface area, facilitating said evaporation, decomposition and hydrolysis, and wherein said impactor comprises hydrolysis catalyst, further facilitating said decomposition and hydrolysis.

15. A selective catalytic reduction device according to claim 2 wherein the material of said impactor is selected from the group consisting of high thermal conductivity and low heat capacity materials.

16. A selective catalytic reduction device according to claim 2 comprising a heat source heating said impactor in addition to heating by said exhaust.

17. A selective catalytic reduction device according to claim 16 wherein said heat source is a voltage source external to said housing, and a pair of electrical conductors connecting said voltage source to said impactor.

18. A selective catalytic reduction device according to claim 2 wherein said impactor has a star shape with a central core and a plurality of radially outwardly projecting tips each providing impaction surface area yet permitting exhaust flow therebetween.

19. A selective catalytic reduction device according to claim 2 wherein said impactor has a bowl shape with a concave surface facing said injector, said concave surface providing increased impaction surface area within a reduced lateral area blocking exhaust flow.

20. A selective catalytic reduction device according to claim 2 wherein said injector has a downstream injection tip, and wherein the distance between said injection tip and said catalyst is less than about 0.5 meters.

21. A method for enhancing ammonia reaction with NO_x in a selective catalytic reduction device for engine exhaust having a housing with an upstream inlet for

receiving said exhaust, and a downstream outlet for discharging said exhaust, comprising injecting a solution comprising a source of ammonia in said housing with an injector to react
5 with and reduce NO_x in said exhaust, providing a catalyst in said housing downstream of said injector, inertially impacting said exhaust and said solution against an inertial impactor in said housing between said injector and said catalyst.

22. A method according to claim 21 comprising holding said solution at said impactor to enhance ammonia reaction with NO_x.

23. A method according to claim 22 comprising providing said source of ammonia as an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust, and holding said aqueous urea solution at said impactor until evaporated, decomposed and hydrolyzed to ammonia.

24. A method according to claim 22 comprising providing said source of ammonia as an ammonia solution, and holding said ammonia solution at said impactor until evaporated.

25. A method according to claim 22 comprising providing a nonporous said impactor, and injecting said solution from said injector at an injection rate less than the solution evaporation rate at said impactor.

26. A method according to claim 25 comprising providing said impactor with a large enough size such that said evaporation rate at said impactor is greater than said injection rate at said injector.

27. A method according to claim 25 comprising providing said source of ammonia as an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust, and injecting said aqueous urea solution from said injector at an injection rate less than the aqueous urea solution
5 evaporation and decomposition rate at said impactor, such that said impactor holds said aqueous urea solution until evaporated and decomposed to ammonia.

28. A method according to claim 22 comprising providing a porous said impactor having a rough porous collection surface, and inertially impacting said solution against said rough porous collection surface causing liquid particle separation of smaller size liquid particles than a smooth nonporous impactor impingement surface to improve
5 evaporation of said solution.

29. A method according to claim 28 comprising wetting said solution to said rough porous collection surface to spread said solution over a greater surface area, facilitating evaporation.

30. A method according to claim 28 comprising providing said source of ammonia as an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust, wetting said aqueous urea solution at said rough porous collection surface such that aqueous urea solution is drawn
5 into said impactor and spread over a greater surface area, facilitating evaporation, decomposition and hydrolysis.

31. A method according to claim 22 comprising providing said source of ammonia as an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust, and catalyzing said aqueous urea solution at said impactor with hydrolysis catalyst, facilitating said decomposition and
5 hydrolysis.

32. A method according to claim 22 comprising selecting the material of said impactor from the group consisting of high thermal conductivity and low heat capacity materials.

33. A method according to claim 22 comprising heating said impactor with a heat source, in addition to heating by said exhaust.

34. A selective catalytic reduction device for engine exhaust comprising:

a housing having an upstream inlet for receiving said exhaust, and a downstream outlet for discharging said exhaust;

an injector in said housing for injecting an aqueous urea solution which
5 evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust;

an SCR catalyst in said housing downstream of said injector;

an impactor in said housing and spaced downstream from said injector and spaced upstream from said SCR catalyst, said impactor comprising a hydrolysis catalyst
10 facilitating decomposition and hydrolysis of said aqueous urea solution at said impactor.

35. A selective catalytic reduction device according to claim 34 wherein said housing has a given cross-sectional area at said SCR catalyst, and wherein said hydrolysis catalyst has a cross-sectional area less than said given cross-sectional area.

36. A selective catalytic reduction device according to claim 34 wherein said housing extends axially along an axis and exhaust flows generally axially through said housing, said aqueous urea solution is injected along an injection spray angle θ from said injector, said hydrolysis catalyst has a lateral cross-sectional dimension D transverse to said
5 axis, said injector has a downstream injection tip spaced from said hydrolysis catalyst by a distance d along said axis and having an inner diameter ϕ transverse to said axis, and wherein $D > 2d \tan (\theta/2) + \phi$.

37. A selective catalytic reduction device according to claim 36 wherein said hydrolysis catalyst is selected from the group consisting of porous and fibrous ceramic material.

38. A selective catalytic reduction device according to claim 36 wherein said hydrolysis catalyst is selected from the group consisting of: extruded material; wrapped material; and layered material.

39. A selective catalytic reduction device according to claim 36 wherein said hydrolysis catalyst is selected from the group consisting of: extruded monolith material;

composite ceramic material; fibrous material; and metallic material.

40. A selective catalytic reduction device according to claim 34 comprising a heat source heating said hydrolysis catalyst in addition to heating by said exhaust.

41. A method for reducing the distance between an injector and SCR catalyst and reducing the size of the SCR catalyst and reducing pressure drop and increasing time for ammonia mixing in a selective catalytic reduction system having a selective catalytic reduction device for engine exhaust having a housing with an upstream inlet for receiving said exhaust, and a downstream outlet for discharging said exhaust, an injector injecting an aqueous urea solution which evaporates, decomposes and hydrolyzes to produce ammonia to react with and reduce NO_x in said exhaust, and an SCR catalyst in said housing downstream of said injector, said method comprising inertially impacting said exhaust and said aqueous urea solution against a hydrolysis catalyst impactor in said housing spaced downstream from said injector and spaced upstream from said SCR catalyst, said housing having a given cross-sectional area at said SCR catalyst, said hydrolysis catalyst having a cross-sectional area less than said given cross-sectional area.

42. A selective catalytic reduction device substantially as described hereinbefore, with reference to the accompanying drawings and/or as shown in Figure 1, Figure 2 and Figure 7; or Figure 1, Figure 3 and Figure 7; or Figure 1, Figure 5, Figure 6 and Figure 7; or Figure 1, Figure 4 and Figure 7; or Figure 8 and Figure 9.

43. A method for enhancing ammonia reaction with NO_x substantially as described hereinbefore with reference to the accompanying drawings.

44. A method for reducing the distance between an injector and SCR catalyst and reducing the size of the SCR catalyst and reducing pressure drop and increasing time for ammonia mixing in a selective catalytic reduction system substantially as described hereinbefore with reference to the accompanying drawings.



INVESTOR IN PEOPLE

Application No: GB 0224218.8
Claims searched: 1-44

Examiner: Dr Albert Mthupha
Date of search: 23 April 2003

Patents Act 1977 : Search Report under Section 17

Documents considered to be relevant:

Category	Relevant to claims	Identity of document and passage or figure of particular relevance	
X	1, 2, 3, 4, 10, 13, 14, 21, 22, 23, 34, 41 at least.	US 5643536 A	SIEMENS, see column 4 line 57-column 5 line 18, Figures 1-4.
X	1, 2, 3, 4, 10, 13, 14, 21, 22, 23, 34, 41 at least.	DE 4417238 A1	SIEMENS, see whole document, note particularly Figure 1.
X	1, 2, 3, 20 at least.	WO 98/28070 A1	CLEAN DIESEL TECH., see whole document, note particularly page 11 lines 24-27, page 13 lines 24-25, page 16 line 26-page 17 line 5, Claims 1, 4, 7, 8, 11, 13, 15.
A		WPI Abstract AN 1999-413274 [35] & JP 11166410 A (HINO), see Abstract & Figures 1, 2, 3, 4.	

Categories:

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.

Field of Search:

Search of GB, EP, WO & US patent documents classified in the following areas of the UKC^v:

B1W

Worldwide search of patent documents classified in the following areas of the IPC⁷:

B01D; B01J; F01N; F02D

The following online and other databases have been used in the preparation of this search report:

ONLINE : EPODOC, JAPIO, WPI.